July 1, 2014

Characterisation of a stratospheric sulphate plume from the Nabro volcano using a combination of passive satellite measurements in limb and nadir geometry
by M. Penning de Vries et al.

Authors' reply to the interactive comment by anonymous referee 2

We thank the referee for his/her concise review of our manuscript. The referee's comments helped us improve our argumentation, in particular with regard to the effects of ice clouds on our measurements. We agree with most of the referee's comments; our replies and the changes made to the manuscript are given below, colour-coded for clarity:
Green: Referee comment.
Black: Authors' reply.
Red: Modified text in manuscript.

Referee comment: 1. Page 7741: 18-21. This gives the impression that the 'first volcanic plume' was confined to 18-19 km. The cited papers do not confirm does. Depending on the definition of 'first volcanic plume' (Nabro erupted almost uninterrupted for the first two weeks), it would be more correct to say 'above 10 km, with a maximum around 18-19 km.'

Authors' reply: We thank the referee for the good suggestion. On account of this comment and of the first comment by Marc von Hobe, we modified the sentence on lines 15-20 to:

A large fraction of the volcanic plumes was injected below the local tropopause (dynamic tropopause at 16.8 km altitude according to ECMWF), but this was not the case for the plume from the first explosion (and possibly later plumes) from Nabro. In contrast to the analysis in (Bourassa et al., 2012), evidence has emerged that at least parts of the initial volcanic plume (emitted on 12-13 June, and here referred to as the 'first volcanic plume') were emitted at the top of or even above the tropopause and were located above 10 km, with a maximum around 18-19 km within hours of the eruption (Sawamura et al., 2012; Fromm et al., 2013; Vernier et al., 2013; Fairlie et al., 2013; Clarisse et al., 2014).

Referee comment: 2. Page 7741: 26. These 30-40 days for SO_2 lifetime were derived for plumes above 25 km (Pinatubo and El Chichon eruptions). The lifetime is highly dependent on the altitude/water vapour content, and for lower plumes it will be much shorter. For example: Kasatochi (2009): 9 days (Krotkov, N. et al. J. Geophys. Res., 2010, 115, D00L20) Sarychev (2011): 10-14 days (Haywood, J. et al. J. Geophys. Res., 2010, 115, D21212) Cerro Hudson (1991): 6 days (Constantine, E. K. et al., AGU Monograph 116 - Remote Sensing of Active Volcanism, 2000, edited by P. Mouginis-Mark et al., pp. 45-64) For Nabro it was estimated to be around 5 days (Theys, N. et al. Atmos. Chem. Phys., 2013, 13, 5945-5968). In view of this, it is not surprising that stratospheric aerosols can be detected very early on.

Authors' reply: The referee is right in saying that the lifetime of 30-40 days found for Pinatubo and El Chichon is probably not appropriate for Nabro, and that we should have mentioned the life time of Nabro's SO_2 plume determined by Theys and co-workers. We now discuss the issue in more detail and include the suggested references. The section was altered, also to comply with the comments made by Simon Carn, as follows:

This assumption is based on data from eruption plumes at altitudes above 25 km (Pinatubo and El Chichón) and SO_2 life times are much shorter for volcanic plumes at lower altitudes or with higher water vapour content: life times on the order of 1-2 weeks were found for Kasatochi (Krotkov et al., 2010), Sarychev (Haywood et al., 2010), and Cerro Hudson (Constantine et al., 2000). For the earliest emitted "high-altitude" plumes (at 15-18 km) from Nabro, an SO_2 life time of 5 days was found (Theys et al., 2013) and, consequently, an amount of sulphate aerosols large enough to be visible from space could be observed within at most 36 hours of the eruption. This is in agreement with a study by (Sawamura et al., 2012), who report the presence of an aerosol plume in the lower stratosphere at 17 km altitude using data from a ground-based lidar station in Sede Boker,

Israel, approximately 30 hours downwind of the first explosion of Nabro.

Referee comment: 3. Page 7744: Section 2.2. It is discussed in the conclusion of the paper, but it would be very helpful if the effect of water and ice clouds on UVAI (and on the sign) could be described here.

Authors' reply: The effect of ice and water clouds on UVAI is, indeed, an important (and as yet still unclarified) issue for this paper. We decided to move the discussion about the effect of ice on UVAI to the results section (see the answer to comment 5), but do not discuss it in section 2.2 yet. That section has been changed to include more information on UVAI (see our reply to the next comment) and we prefer to restrict it to technical information.

Referee comment: 4. Page 7744: Section 2.2 first paragraph. The description of UVAI, AAI and SCI is confusing. Do all three refer to the same quantity and only differ in the sign? 'The positive part' is confusing, either it is positive or it is negative. Likewise 'the counterpart' is confusing. I would suggest rewriting/expanding this paragraph, starting with a definition of UVAI and then describing the effect of scattering or absorbing aerosols on the index using comprehensive examples (ash, sand, water, smoke, sulfates, trace gases(?) etc..). Why does the SCI not depend on the plume altitude, while the AAI does?

Authors' reply: The referee is right – as it is a point of focus of the manuscript, the description of UVAI deserves more attention. We expanded section 2.2 and included the UVAI equation, as suggested by Simon Carn. We removed the statement 'SCI does not depend on plume altitude', because this is only valid in nadir geometry for aerosols with single-scattering albedo equal to 1.0 (see Fig. S1A in the supplementary material). We do note, however, that the altitude dependence of SCI is smaller than that of AAI. Section 2.2 now reads:

To investigate if ash was present in the volcanic plume we use the UVAI (also known as 'residue'). The definition of the UVAI is based on the principle that aerosol optical properties differ from those of molecules, so that the presence of an aerosol layer leads to a change in contrast between the reflectance at a wavelength λ (e.g. 340 nm) and at a reference wavelength λ_0 (e.g. 380 nm) in comparison to an aerosol-free atmosphere. The calculation of UVAI requires radiative transfer modelling of reflectances at λ and λ_0 for aerosol-free scenes bounded by a Lambertian surface with an albedo value chosen such that measured and modelled reflectances at λ_0 are equal. The UVAI is obtained by calculating the ratio between the measured (R^{meas}) and modelled (R^{model}) reflectances at λ (Torres et al., 1998):

$$UVAI = -100 \cdot log_{10} \left(\frac{R^{meas}}{R^{model}} \right)_{\lambda}$$

Positive values of UVAI are commonly referred to as AAI (Absorbing Aerosol Index), which is a well-known and much-used qualitative indicator of aerosols that absorb radiation in the UV range, such as mineral dust, biomass burning smoke, and volcanic ash (Herman et al., 1997; Torres et al., 1998; de Graaf et al., 2005). The AAI depends on plume altitude and is most sensitive to elevated layers of absorbing aerosols, in particular if these are located over bright surfaces such as ice or clouds (see, e.g. Hsu et al., 1999; Fromm et al., 2010). The SCI (SCattering Index), which consists of UVAI< 0, was only recently introduced as an indicator for aerosols that do not or only weakly absorb UV radiation (Penning de Vries et al., 2009), such as sulphate aerosols and other secondary aerosols formed from anthropogenic or biogenic emissions of trace gases. UVAI generally depends on aerosol amount, size distribution, absorption, and altitude. The AAI is more sensitive to aerosol amount and plume altitude than the SCI because absorption by aerosols causes larger changes in spectral contrast in the UV range than aerosol scattering (Torres et al., 1998; de Graaf et al., 2005; Penning de Vries et al., 2009). A big advantage of the UVAI is that it can be meaningfully interpreted in the presence of clouds; however, clouds influence UVAI and must be taken into account for quantitative analysis of UVAI (Penning de Vries and Wagner, 2011).

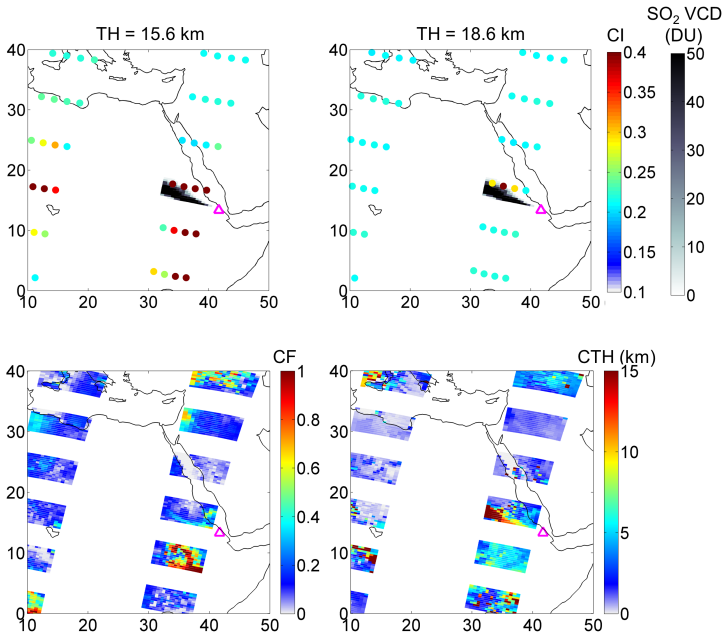
Referee comment: 5. Page 7750: As reported also in several other studies, ice particles were present in great numbers in the Nabro plume on the 13th. As mentioned in the conclusion, they had an impact on the UVAI on the first day - and I think the right place to discuss this is here, and not in the concluding section. (another argument is that there is no reason the UVAI of sulfates alone would be stronger (more negative) on the first day than the second.

Authors' reply: We agree with the referee that the important discussion regarding the effect of (ice) clouds on UVAI should not have been left until the final section of the paper. We rearranged and partly re-wrote the paragraph addressing the influences of ice particles, which now reads:

Most of the particle extinction observed in the MODIS images from 13 June (Fig. 1G and H) is probably due to the presence of ice particles, visible in MODIS IR cloud data (Fromm et al., 2013) and in SEVIRI data (Vernier et al., 2013). On the following day, the plume can no longer be seen in MODIS true-colour images (Fig. 2G and H) and neither is a cloud detected by the IR retrieval (not shown here, but see: http://ladsweb.nascom.nasa.gov/browse_images/granules.html). This does not exclude the possibility that ice was present in the volcanic plume after June 13, but the low depolarization ratio detected by CALIOP is evidence that it consisted mainly of sulphate aerosols (Vernier et al., 2013; Fairlie et al., 2013, Clarisse et al., 2014). In addition, the overpass of the volcanic plume over the joint MPLNET/AERONET (MicroPulse Lidar NETwork (Welton et al., 2001) and AErosol RObotic NETwork (Holben et al., 1998)) site Sede Boker corresponded to a large increase in the number of fine particles (of which the size distribution peaked at approximately 0.1 μm, see: http://aeronet.gsfc.nasa.gov), also clearly indicating that the plume was made up of sulphate aerosols; the optical depth assigned to coarse-mode particles is very similar to that measured on the days before the eruption and can confidently be attributed to mineral dust. Based on the evidence presented above, we conclude that ice contributed substantially to the negative UVAI signal detected on June 13, but that the UVAI signal on June 14 was caused by sulphate aerosols.

Referee comment: 6. Figure 4. Why is this shown at 18.6 km (while the profile maxima are at 13.5 and 16.5 km respectively)?

Authors' reply: The figure was included to give an overview of the situation using the qualitative Colour Index (CI). As the referee correctly notes, the extinction due to the volcanic plume is maximal at 13.5 km (June 13) and 16.5 km (June 14), but at these altitudes the contribution of other scatterers (e.g. cirrus clouds) to the CI is quite large. This can be seen in the upper left panel of the figure below, where we depict the CI at 15.6 km for June 13; the upper right panel shows the CI at 18.6 km for reference. Evidence that most of the enhanced CI at 15.6 km are caused by clouds comes from the (nadir) FRESCO data presented in the lower left (effective cloud fraction, CF) and right (cloud top height, CTH) panels: enhanced CI values are often found in pixels with mid- to high-level clouds (CTH> 5 km). Some of the enhanced CI values are probably caused by smaller or thinner clouds (e.g. cirrus) which may be missed by the FRESCO algorithm. Note that the volcanic plume appears as a high cloud in the FRESCO data.



Upper row: maps of the SCIAMACHY Colour Index at tangent heights of 15.6 km (left) and at 18.6 km (right) for the region around the Nabro volcano on 13 June 2011. SO_2 columns from SCIAMACHY are shown in the background with a grey colour scale. The volcano is marked by a pink triangle (compare Fig. 5 in the manuscript). Lower row: FRESCO effective cloud fraction (left) and cloud top height (right) for SCIAMACHY nadir data from June 13, 2011.

Since the volcanic plume shows clearly enhanced extinction with respect to background conditions even at 18.6 km (see Figure 5), whereas the contribution from clouds is nearly negligible, this tangent height was chosen for the CI in Figure 4. We added this information to the text by changing the sentence starting on line 19 to:

Each dot is colour-coded by the CI at a TH of 18.6 km, which is clearly above the tropopause (at 16.8 km, according to ECMWF) and is therefore largely unaffected by other scatterers, e.g. cirrus clouds.

Referee comment: 7. Figure 5. The profiles shown in figure 5 are very broad (≈ 10 km), surely broader than the actual plume. Could you comment on why this is the case?

Authors' reply: The broadness of the retrieved plumes is a result of the coarse vertical resolution of SCIAMACHY, i.e. 3 km. In the worst case, if the centre of an aerosol layer is located at the border between two tangent heights, no matter how thin the layer, a similar amount of aerosols (depending on the instrument's sensitivity) is retrieved at both altitudes, resulting in an apparent plume thickness of 6 km. This worse case scenario appears to be very close to the situation encountered here, with a plume at 16-17 km and tangent points at 15.6 and 18.6 km. The profile is additionally broadened due to the so-called 3D effect, i.e., the plume is not infinite and not centred at the TP. The fact that the limb data are interpolated onto a constant altitude grid with steps of 3 km (from a temporally changing grid with 3.3 km spacing) prior to the retrieval also causes broadening. We tried to make this more apparent by adding bars indicating SCIAMACHY's resolution to Figs. 5 and 9. As an example, we show the updated Fig. 5 and caption here:

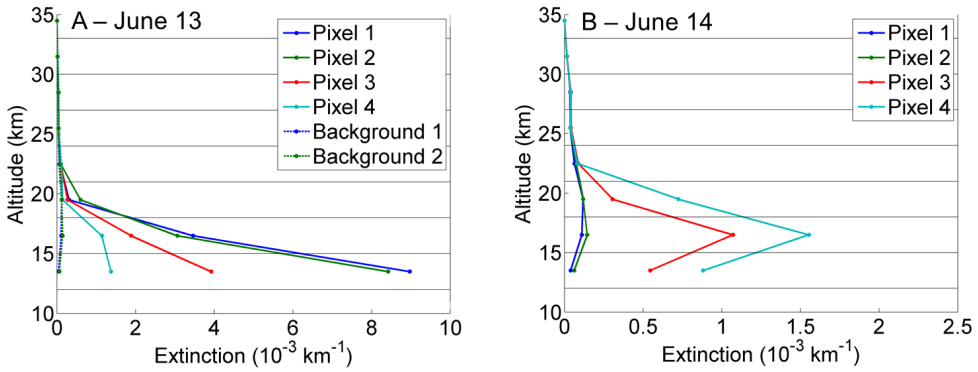


Fig. 5. Limb extinction profiles of all limb pixels in the SCIAMACHY state containing parts of the SO_2 plume on June 13 (state 20 of orbit 48555; panel A) and on June 14, 2011 (state 19 of orbit 48570; panel B). The resolution of the instrument is indicated by the black grid lines. The two profiles labelled "Background" (dashed lines in panel A) are the profiles from pixels 1 and 2 in on June 14 (panel B). Note that the panels have different x-axis scales. Pixel enumeration is from West to East.

We also added a statement addressing the broadness of the retrieved plume to Sect. 4.2, which reads:

Note that the 3-km vertical resolution of SCIAMACHY (along with 3D effects that will be introduced shortly) causes the retrieved aerosol layer to appear much broader than the actual plume.

Referee comment: 8. Figure 6. Why was the aerosol layer put at such high altitudes? 16-19 km would be more in line with what was actually observed in that part of the Nabro plume.

Authors' reply: The referee is right, and we changed the caption and the accompanying Fig. 7 for consistency. Note however, that Fig. 6 itself does not change if the aerosol layer is put at another altitude.

Referee comment: 9. Figure 7. The effect of a plume not centred at the tangent point is very interesting. Have any attempts been made to correct for this effect in the retrieval using collocated SO_2 nadir retrievals?

Authors' reply: We absolutely agree with the referee that it is an interesting finding. We made some attempts at correcting the limb retrieval using nadir SO_2 data as a proxy for the location of the aerosol plume. However, resolving this issue is more difficult than it appears, e.g. due to the fact that background aerosols

need to be retrieved simultaneously, but are not confined to the volcanic plume. We therefore decided not to include this into the current paper. We are investigating these so-called 3D effects in more detail and plan to publish the results and a more detailed description of the algorithm in a manuscript dedicated to our new limb retrieval; an earlier version appeared in the 2013 ESA conference proceedings: Dörner et al.: Retrieval of stratospheric aerosol properties from SCIAMACHY limb observations, in: Proc. ESA conference ATMOS 2013, SP-482). We added the following sentence to page 7753, line 7:

These effects cannot easily be corrected for in our retrieval algorithm due to, e.g., the necessity to define a non-localized background aerosol in addition to the localized volcanic aerosol in the retrieval. We are investigating these so-called 3D effects in more detail and will publish the results and a more detailed description of the algorithm in a manuscript dedicated to the limb retrieval; an earlier version was presented in (Dörner et al., 2013).

Referee comment: 10. Page 7753: 14-22. this part is not very clear. Also line 17, the comment on pixel 3 and 4, does not seem to be consistent with what is shown in the figure.

Authors' reply: We tried to improve the readability by changing lines 14-22 to:

The SCIAMACHY limb retrieval detects the ice cloud at slightly lower altitude (possibly due to the fact that the cloud is not centred at the TP) and is of such high optical density that the limb retrievals below the maximum (at 16.5 km) are affected and probably yield extinctions that are unrealistically high. Only for optically very thin aerosol layers, such as the volcanic plume shown in panel 5B, both plume top and bottom can be retrieved. On the other hand, lower-lying aerosols (such as those detected by the MPLNET lidar at Sede Boker) may also be the cause of the increased extinction at 13.5 km in panel A.

Lines 17-22 were removed, as the statements regarding the 3D effects on the profiles presented in Fig. 5B are, as the referee notes, not very apparent.

Referee comment: 11. Page 7753: 27. there was no 'merging of plumes'; the early plume was continuous but slanted over an altitude range. What is seen in subsequent days (16/17 June) is a horizontal separation of the plume due to differential wind shear.

Authors' reply: This was clearly an unfortunate choice of wording. We did not mean that the plumes truly merged (and therefore called it 'apparent merging'), but we admit that this was rather confusing. The sentence was changed to:

Although the SO_2 plumes detected on June 15 overlap in the horizontal plane (best seen in panel A between $40° - 70°$E), the difference in plume altitude and resulting differences in wind speed and direction ensure that the two plumes can be observed separately again on June 16 and 17.

Referee comment: 12. Page 7755: 3-7. As far as I can tell, this is not seen in the figures. Please check and correct/clarify.

Authors' reply: The downward shift and the underestimation is best seen in the highest part of the plume: the extinction retrieved at 19.5 km is clearly enhanced in pixels 2 and 3 with respect to pixels 1 and 4. This was clarified in the text by changing the sentence starting on line 3 to:

The plume is located further from the TPs of pixels 1 and 4, which translates in a downward shift of the retrieved profiles relative to those of pixels 2 and 3: the retrieved extinction at 19.5 km is significantly higher in pixels 2 and 3.

Referee comment: 13. Page 7758: 27: 17-20 km is not in agreement with the presented results. 15-20 km would be more in line with what is actually shown.

Authors' reply: The referee is right, the extinction maximum almost always appears in the 3-km high box centred at 16.5 km, so the correct range is 15-20 km. We corrected the error.